

THz spectroscopy of the atmosphere

Herbert M. Pickett^a

Jet Propulsion Laboratory, California Institute of Technology,
Mail Stop 183-701, Pasadena, CA 91109

ABSTRACT

THz spectroscopy of the atmosphere has been driven by the need to make remote sensing measurements of OH. While the THz region can be used for sensitive detection on many atmospheric molecules, the THz region is the best region for measuring the diurnal behavior of stratospheric OH by remote sensing. The infrared region near 3 μm suffers from chemiluminescence and from spectral contamination due to water. The ultraviolet region near 300 nm requires solar illumination. The three techniques for OH emission measurements in the THz region include Fourier Transform interferometry, Fabry-Perot interferometry, and heterodyne radiometry. The first two use cryogenic direct detectors while the last technique uses a local oscillator and a mixer to down convert the THz signal to GHz frequencies. All techniques have been used to measure stratospheric OH from balloon platforms. OH results from the Fabry-Perot based FILOS instrument will be given. Heterodyne measurement of OH at 2.5 THz has been selected to be a component of the Microwave Limb Sounder on the Earth Observing System CHEM-1 polar satellite. The design of this instrument will be described. A balloon-based prototype heterodyne 2.5 THz radiometer had its first flight on 24 May 1998. Results from this flight will be presented.

Keywords: THz, atmosphere, stratosphere, heterodyne

1. INTRODUCTION

Stratospheric ozone depletion is controlled by four families of catalytic cycles. In the middle stratosphere (28-35 km), the NO / NO₂ pair dominates ozone depletion in the natural atmosphere. When anthropogenic chlorofluorocarbons are present, the Cl / ClO pair also contributes significantly to ozone depletion in the middle stratosphere. In the region of 40-60 km, the OH / HO₂ pair dominates ozone depletion, while above this altitude the H / OH pair is the dominant contributor. Recently, it has been recognized that heterogeneous chemistry on aerosols plays an important role in reducing the concentration of nitrogen radical and in increasing the amount of nitric acid. Both effects of aerosols can make the OH / HO₂ pair dominant below ~28 km. While NO₂ has been measured from space in the infrared and ultraviolet and ClO has been measured from space at millimeter wavelengths, stratospheric OH has not been measured from space at altitudes below 50 km.

1.1. Measurement techniques

THz spectroscopy of the atmosphere has been driven by the need to make remote sensing measurements of OH. While the THz region can be used for sensitive detection on many atmospheric molecules, the THz region is the best region for measuring the diurnal behavior of stratospheric OH by remote sensing. The OH measurement in the infrared region near 3 μm suffers from chemiluminescence and from spectral contamination due to water. The ultraviolet region near 300 nm requires solar illumination and suffers from absorption by atmospheric ozone. The three techniques for OH emission measurements in the THz region include Fourier transform spectrometers (FTS), Fabry-Perot interferometry, and heterodyne radiometry. The first two use cryogenic direct detectors while the last technique uses a local oscillator and a mixer to down convert the THz signal to GHz frequencies.

All three techniques have been used to measure stratospheric OH from balloon platforms. FTS instruments have been developed by Harvard Smithsonian Astrophysical Observatory¹ and by a collaboration of IROE (Firenze, Italy) and NASA Langley Research Center.² Both instruments are quite large and have high data rates, but have shown excellent capability for sensitive measurements of a number of stratospheric molecules in addition to OH. The JPL Far Infrared Limb Observing Spectrometer (FILOS) is an example of a Fabry-Perot based instrument.³ It is much smaller, has a low data rate, and is designed specifically to measure OH. Heterodyne measurement of OH at 2.5 THz has been selected to be a component of the Microwave Limb Sounder on the Earth Observing System CHEM-1 polar satellite. Further details are given below. A balloon-based prototype heterodyne 2.5 THz radiometer (BOH) had its first flight on 24 May 1998, and flew jointly with FILOS. Preliminary results from this flight are given below.

^a Correspondence: hmp@spec.jpl.nasa.gov; Telephone: 818 354 6861; Fax: 818 393 5065

The FILOS instrument has been part of a total of 14 balloon flights. The modest requirements of FILOS have allowed it to fly with either of the FTS instruments as well as the new the new BOH instrument. In effect, the FILOS instrument can serve as a transfer standard between different instruments that could not otherwise be compared. A detailed discussion of the results of these comparison flights will be part of a future paper, but initial comparisons show excellent agreement between FILOS and the FTS derived profiles of OH. An earlier paper has shown that the FILOS-measured OH profiles and diurnal behavior are consistent with simple photochemical models the utilize measured ozone and water profiles.⁴

2.EOS MLS 2.5 THz OH CHANNEL

Heterodyne measurement of OH at 2.5 THz has been selected to be a key component of the Microwave Limb Sounder on the Earth Observing System CHEM-1 polar satellite. The requirements of this channel are to obtain monthly global maps with 5° latitude resolution and 3 km height resolution from 18 to 60 km. Projected sensitivity for high altitude (45-60 km) OH will allow daily maps, but more integration is required to obtain maps down to 18 km. It is not practical to look lower than 18 km because the water and dry air continua attenuate the OH signal for tangent heights below this altitude. As can be seen from Figure 1, OH has a very string gradient in volume mixing ratio, dropping below the parts-per-trillion level at 20 km. The projected sensitivity is also shown based on a $T_{\text{sys}} = 30\,000\text{ K}$ (SSB) using two OH polarizations. This sensitivity is possible in part because of the limb sounding geometry and in part because the line shape is resolved.

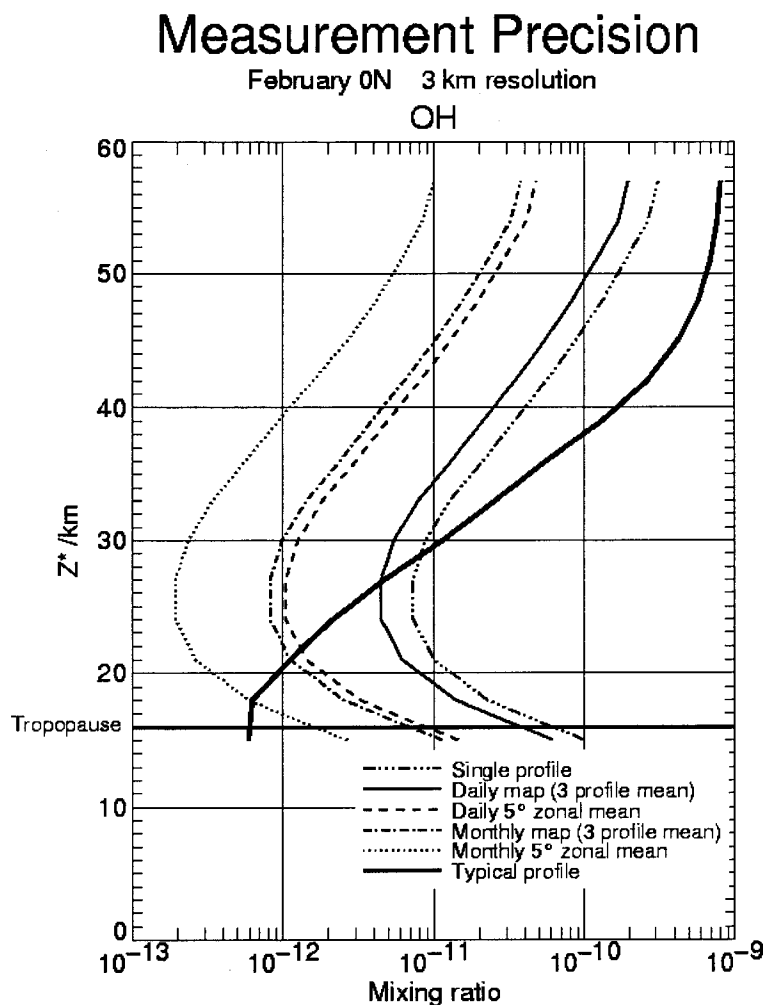


Figure 1: EOS-MLS OH Sensitivity

The steep gradient of concentration means that extremely accurate pointing information is required. In fact, to obtain the sensitivity needed for the monthly maps pointing accuracy of 100 m is needed. Fortunately, nature placed an oxygen emission line within 8 GHz of the OH lines, and this emission line will be used to register the scan on the atmosphere. Temperature profiles will be measured by the lower frequency channels and will be used to calculate radiance of the THz oxygen line with respect to scan angle. Comparison of the offset between calculation and observation then establishes to pointing offset.

At first, the OH radiometer was to share the main 1600×800 cm main antenna, but the high frequency drove antenna requirements. In addition, the THz channel imposed constraints on system test and calibration because the THz channel performance can only be adequately measured in a vacuum, while the other four radiometers can be tested in air. The THz channel has a separate antenna and scanning system as shown in Figure 2. However, it still shares the same filter bank and data system. The antenna is a 22.8 cm offset Gregorian telescope with a 26.4×37.3 cm elliptical scanning mirror in front of the telescope. The scan mirror can be directed toward the limb, to a cold view above the atmosphere, or at an ambient temperature load.

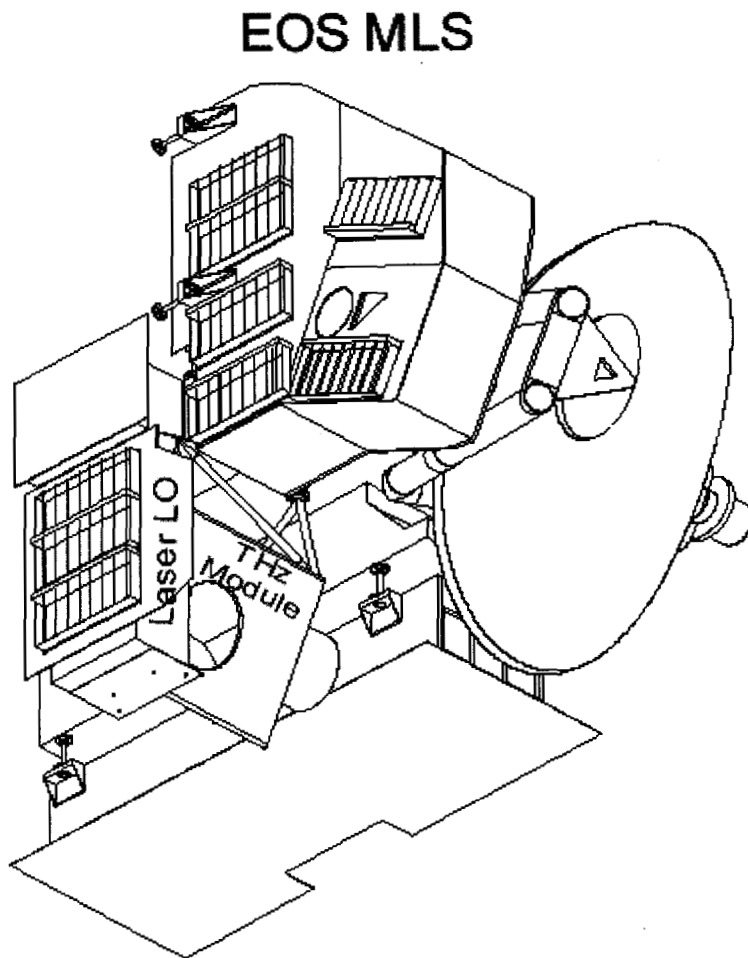


Figure 2: EOS-MLS Modules

The radiometer consists of a laser local oscillator, a quasi-optical diplexer, and two mixers that are oriented to receive horizontal and vertical polarizations from the atmosphere. The atmospheric radiance is not expected to be significantly polarized and the two mixer signals are only used to improve the S/N and provide some redundancy. The local oscillator is a methanol laser pumped by a CO₂ laser. The laser is being provided by DeMaria Electro Optics Systems. It will have 20 mW output power for 120 W input at 28 V. Currently we are pursuing two mixer approaches. The first is a whisker-contacted

waveguide mixer being developed by Rutherford Appleton Lab⁵ and the second is a membrane-supported planar waveguide mixer developed at the Jet Propulsion Lab.⁶

3. FIRST BALLOON HETERODYNE OH MEASUREMENTS

A balloon-based prototype heterodyne 2.5 THz radiometer had its first flight on 24 May 1998 from Ft. Sumner, NM. The purpose of this balloon instrument is to provide early real-world use of selected components that will be used on the flight instruments, to obtain early views of stratospheric OH using the frequencies and techniques that will be used in flight, and to gain operational experience with a balloon instrument that can be used for sub-orbital validation after launch. The balloon instrument included a JPL waveguide mixer, a prototype laser LO, and a brassboard filter bank. The second LO frequencies were nearly the same as specified for EOS MLS except that a correction for the 56 MHz spacecraft Doppler shift was not included. The mixer, laser LO, and the filter bank worked well during the flight, but a coolant pump failed causing the thermal control to be much worse than desired. Fortunately, for a period of 40 min midway through the flight, the thermal drift was small enough to lock the laser and take data. Results for this flight are given in Figure 3. The solid line is the calculated OH emission after fitting the limb scans to the data. The fitted profile of OH is quite similar to values previously obtained from Ft. Sumner.

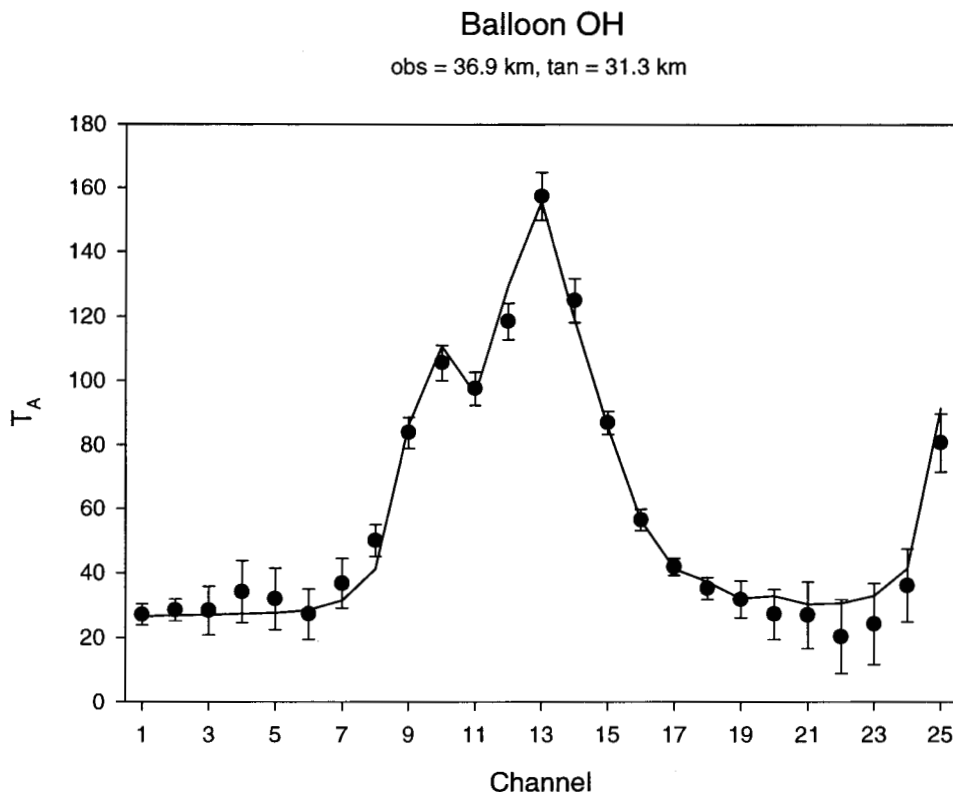


Figure 3: OH Stratospheric Emission from Balloon

4. CONCLUSIONS AND FUTURE PROSPECTS

Heterodyne techniques in the Terahertz region will provide significant improvement in our capability to measure OH in the stratosphere. Our balloon instrument has demonstrated the basic capability for such measurements. We plan to fly the balloon instrument again in September 1999 so that the sensitivity can be verified. We also plan to fly FILOS on the same payload so that the OH measured by the two techniques can be compared. Of course, the real insight into stratospheric OH chemistry will come in 2002 when EOS-MLS is launched. The design lifetime of all the CHEM-1 instruments is 5 years, and we expect that a great deal will be learned about the stratosphere and upper troposphere during that observational period.

Present improvements in THz mixer and local oscillator technology will enable improved sensitivity for OH and will extend heterodyne techniques to other molecules in the stratosphere. With a frequency-agile high-sensitivity THz heterodyne receiver, instruments could be developed for space that can respond quickly to changing needs in atmospheric chemistry while retaining superb detection capabilities.

ACKNOWLEDGEMENTS

This work was carried out under contract between California Institute of Technology and the National Aeronautics and Space Administration. I also thank T. L. Crawford and J. C. Pearson for assistance with the May 1998 balloon flight. I also thank the members of the Microwave Limb Sounder team who have helped with the THz channel on EOS or have provided components for BOH.

REFERENCES

1. W. A. Traub, D. G. Johnson, and K. V. Chance, "Stratospheric Hydroperoxyl Measurements," *Science*, vol. 247, 446-449 (1990).
2. B. Carli, M. Carlotti, B. M. Dinelli, F. Mencarglia, and J. H. Park, "The Mixing Ratio of the Stratospheric Hydroxyl Radical from Far Infrared Emission Measurements," *J. Geophys. Res.*, vol. 94, 11049-11058 (1989).
3. H. M. Pickett, and D. B. Peterson, "Far-IR Fabry-Perot Spectrometer for OH Measurements," *SPIE Optical Methods in Atmospheric Chemistry*, vol. 1715, 451-456 (1992).
4. H. M. Pickett, and D. B. Peterson, "Comparison of Measured Stratospheric OH with Prediction," *J. Geophys. Res.*, vol. 101, 16789-16796 (1996).
5. B. N. Ellison, B. J. Maddison, C. M. Mann, D. N. Matheson, M. L. Oldfield, S. Marazita, T. W. Crowe, P. Maaskant, and W. M. Kelly, "First Results for a 2.5 THz Schottky Diode Waveguide Mixer," *Proc. of the 7th Intl. Symposium on Space THz Tech.*, Charlottesville, VA, 494-502 (1996).
6. P. H. Siegel, R. P. Smith, M. Gaidis, S. C. Martin, and J. Podosek, "2.5 THz GaAs Monolithic Membrane-Diode Mixer," *Proc. of the 9th Int'l Symposium on Space THz Tech.*, Pasadena, CA, 147-159 (1998).